Using the ORIGEN2 Code to Resolve Isobaric Interferences in ICPMS Analysis of Spent Fuel

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Introduction

The Chemical Engineering Division's work on separation of spent nuclear fuel required the analysis of many samples of a broad range of elements. The Analytical Chemistry Laboratory has an inductively coupled plasma mass spectrometer (ICP/MS) in a controlled area that was able to handle the sample load and provide fast turnaround of results. Interpreting the data produced from the analysis of spent fuel by ICP/MS requires correction for isobaric effects because the sample material is separated by mass and not by element.

A scheme was developed to use the ORIGEN2 code prediction of fuel rod composition to correct for isobaric effects and provide a comprehensive analysis covering much of the periodic table.

What is the ORIGEN2 Code?

ORIGEN2 is a computer code designed to mathematically calculate the composition of nuclear materials as a function of decay, burnup, initial enrichment and reactor type.

What are isobars and isobaric effects?

Isobars are isotopes of different elements that have the same mass number (e.g., U-238 and Pu-238).

When a sample containing isobars is separated by mass, the resulting data contain the sum of all isobars (elements) within that particular mass.

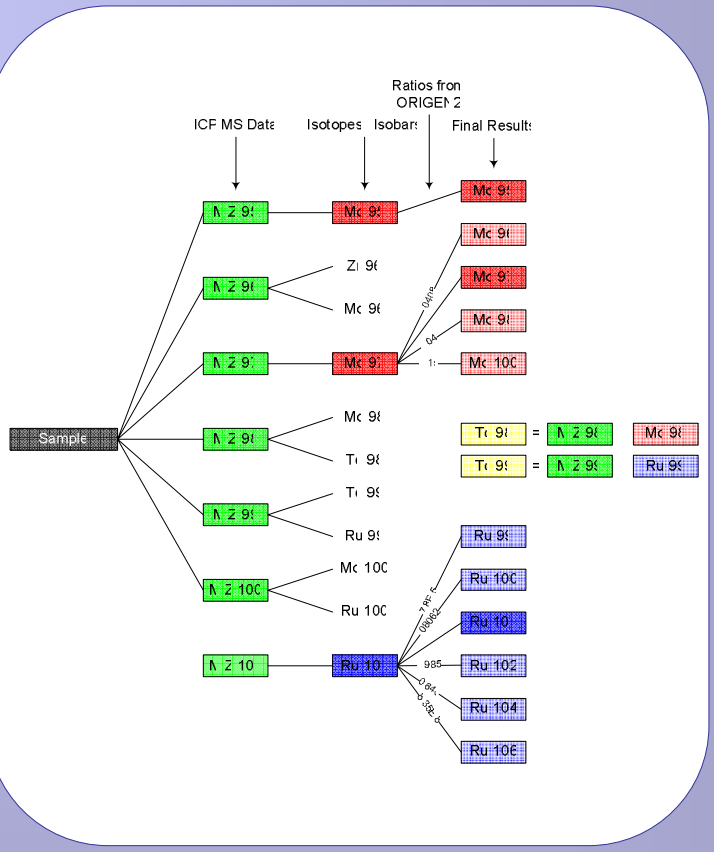
Decoupling ICP/MS results using the ORIGEN2 Code

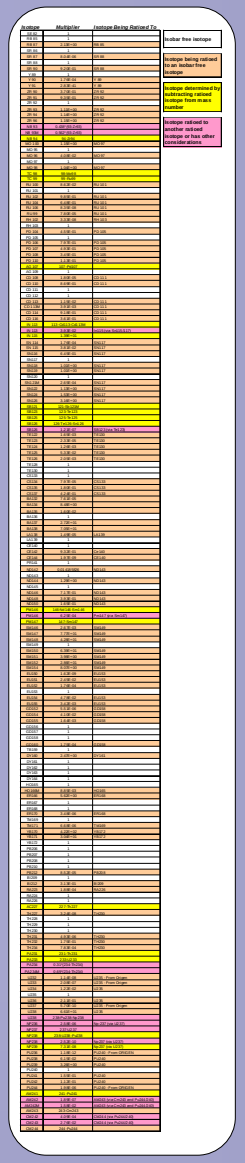
The ORIGEN2 code predictions for the fuel composition were used for all isotopes containing less than 10^{-10} grams (in the ORIGEN2 code output) starting with mass 82. Special notice was made of masses with only one predicted isotope (no isobars). All other expected isotopes of that element were ratioed to the isobar free isotope. In the cases where there were no isobar free isotopes of a particular element, other isotopes of the same given mass (calculated via ratios) were subtracted from the ICP/MS results for that mass.

Confirming the identity of fuel samples

In order to estimate the accuracy of the ORIGEN2 calculations, we separated the uranium and plutonium and analyzed them for their isotopic ratios via TIMS. It was noticed that the ratios differed to those predicted by ORIGEN2. The identity of the fuel received was then questioned. When the thermal ionization mass spectroscopy (TIMS) results were compared with ORIGEN2 predictions of other fuel pins, we were able to establish our fuel identity.

	Ratio to U-235		
Isotope	*TIMS*	*ORIGEN Pin 46*	*ORIGEN Pin 7*
U-234	**0.0132**	*0.0180*	**0.0132**
U-236	**0.2297**	*0.3818*	**0.2310**
U-238	**42.9563**	*64.5967*	**46.0919**





Results

The ORIGEN2 code has subsequently been used on other analytical schemes involving irradiated samples. It has been used to help interpret ICP/MS data, assist in the selection of analysis methods, and to aid in the preparation of sample dilutions.

It has been especially useful in this analysis scheme because it allowed a large number of samples to be analyzed for an entire suite of elements economically and efficiently.

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